Pt-Pd MINERALS FROM THE UPPER ZONE OF THE EASTERN BUSHVELD COMPLEX, SOUTH AFRICA

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Abstract

Although the upper zone of the eastern Bushveld Complex is generally known to contain concentrations of the platinum-group elements only in the low-ppb range, samples from a sulfide-enriched anorthosite in the footwall of lower magnetite layer 2 were found to contain several platinum-group minerals (PGM). Detailed microscopic and microprobe analyses revealed the presence of the Pt-Pd-Bi-Te minerals michenerite, moncheite, merenskyite and froodite, as well as a little sperrylite. In all Pt-Pd-Bi-Te minerals, 1:1 substitution between Pt and Pd, as well as between Te and Bi, confirms the well-documented partial solid-solution series between merenskyite and michenerite, between moncheite and insizwaite, and the complete solidsolution series between moncheite and merenskyite. The textural association of the PGM with silicates rather than sulfides, the lateral restriction of the mineralization to a few meters, and the extensive alteration of silicates, sulfides and titaniferous magnetite in some samples are interpreted as evidence for the formation of the PGM from hydrothermal fluids. The upper and lower limits in temperature for these fluids, as derived from the stabilities of michenerite and plagioclase, are in the order of 490° and 400°C, respectively.

Keywords: Pt-Pd mineralization, michenerite, moncheite, merenskyite, froodite, hydrothermal origin, upper zone, Bushveld Complex, South Africa.

SOMMAIRE

Quoique la zone supérieure du secteur Est du complexe du Bushveld, en Afrique du Sud, contient de très faibles teneurs (quelques ppb) en éléments du groupe du platine, en général, nous avons découvert plusieurs minéraux du groupe du platine (MGP) dans des échantillons d'anorthosite enrichie en sulfures, située au dessous du deuxième niveau de magnétite. Les analyses détaillées au microscope et à la microsonde électronique ont révélé la présence de michenerite, monchéite, merenskyite et froodite, minéraux du système Pt-Pd-Bi-Te, de même qu'un peu de sperrylite. Dans tous les MGP de ce système, le Pt se substitue au Pd, et le Te au Bi, ce qui confirme la solution solide partielle bien établie entre merenskyite et michenerite, et entre monchéite et insizwaite, et la solution solide complète qui existe entre monchéite et merenskyite. L'association des MGP avec les silicates plutôt qu'avec les sulfures, la présence de minéralisation sur à peine quelques mètres, et l'altération intense des silicates, sulfures et magnétite titanifère dans certains échantillons témoigneraient de l'importance

d'une phase hydrothermale dans la formation des *MGP*. Cette phase fluide aurait été importante entre 490° et 400°C, d'après les champs de stabilité de la michenerite et du plagioclase, respectivement.

(Traduit par la Rédaction)

Mots-clés: minéralisation en Pt-Pd, michenerite, monchéite, merenskyite, froodite, origine hydrothermale, zone supérieure, complexe du Bushveld, Afrique du Sud.

INTRODUCTION

The upper zone of the Bushveld Igneous Complex consists generally of Fe-rich differentiates and is widely known for its layers of V-bearing magnetite (Von Gruenewaldt et al. 1985). However, it is widely considered to be depleted in platinum-group elements (PGE). A recent survey through the lower part of the upper zone in the eastern Bushveld Complex showed that this stratigraphic sequence generally contains PGE concentrations only in the low-ppb range (Harney et al. 1990). This observation is in general agreement with previous investigations reported by Page et al. (1982). However, unusually enriched samples with up to 2 ppm total PGE were encountered by Von Gruenewaldt (1976) and Page et al. (1982) from a sulfide-bearing anorthosite in the footwall of the lower magnetite layer 2 (LL2; nomenclature after Molyneux 1970). In the framework of their investigation, Harney et al. (1990) resampled this horizon and found less than 50 ppb PGE in each of four split samples taken from the same locality as the PGE-rich samples encountered by Page et al. (1982).

To investigate the reasons for this disagreement in the analytical data, the anorthosite below LL2 was again resampled, but this time at precisely the same locality as that from which Von Gruenewaldt (1976) previously obtained the *PGE*-enriched material. Microscopic investigations of these samples revealed the presence of several platinum-group minerals (*PGM*), mainly merenskyite, michenerite and moncheite. The aim of this investigation is to document the nature and composition of the *PGM* present, and to explain this locally very restricted concentration of the *PGE*.



FIG. 1. Geological map of the eastern Bushveld Complex in the vicinity of Roossenekal (modified after Visser 1984), showing sample location.

SAMPLING AND MINERALOGICAL DESCRIPTION

Samples were taken from a surface exposure of the upper zone along the Meetse – Matau River on the farm Zwartkop, about 10 km north of Roossenekal in the eastern Bushveld Complex. The general geology and stratigraphy in this part of the intrusion are summarized in Figures 1 and 2. In this area, the upper zone consists predominantly of magnetite-bearing gabbronorites, with interlayered anorthosites and magnetite layers. A detailed description of the geology in the Roossenekal area was given by Von Gruenewaldt (1973). The geology of the Meetse – Matau River section was described by Hammerbeck (1965).

In the locality investigated, the footwall of LL2 consists mainly of an anorthosite that contains, in addition to plagioclase, minor amounts of clinopyroxene, titaniferous magnetite and sulfide. However, the modal composition of the anorthosite varies along strike and can become gabbronoritic, in which case the rock contains up to 40 vol.% clinopyroxene and inverted pigeonite. The sulfide assemblage consists, in decreasing order of abundance, of chalcopyrite, pyrrhotite, pentlandite and pyrite, as well as minor amounts of violarite, sphalerite, galena, mackinawite, marcasite, argentian pentlandite, cobaltite, covellite and the PGM. The total sulfide concentration is variable on a thinsection scale, and ranges from traces up to 7 vol.%, though most samples contain 3 vol.% or more.

The samples display variable signs of alteration. These manifestations include alteration of plagioclase to calcite, quartz, chlorite and sericite, alteration of pyroxene to hornblende, chlorite, calcite and quartz, and replacement of pentlandite and pyrrhotite by violarite and marcasite, respectively. Furthermore, titaniferous magnetite is replaced by a very finegrained intergrowth of secondary silicates, which leaves a network of ilmenite lamellae. The ilmenite is more resistant to replacement than the host magnetite. Not all these alterations are visible in all sections under investigation. However, they occur regardless of the amount of sulfides or PGM present in the section. Hence, no systematic difference in the style of alteration between samples with and without PGM was observed.

ANALYTICAL TECHNIQUES

Quantitative analyses of merenskyite, michenerite, moncheite and froodite were performed at the University of Pretoria using a fully automated JEOL 733 SUPERPROBE with four spectrometers in wavelength-dispersion mode. Owing to the very small size of the *PGM*, quantitative analyses could not be obtained for all grains observed in the samples. The spatial resolution of an electron beam depends on



FIG. 2. Generalized columnar section of the lower part of the upper zone in the Roossenekal area, eastern Bushveld Complex (modified after Von Gruenewaldt 1973). Arrow indicates the stratigraphic position of the samples. Height in meters relative to the base of the main magnetite layer.

the density of the mineral, the accelerating potential and the excitation energy of the element-specific X-ray line. With the formula given by Reed (1975), the spatial resolution was calculated for all elements present at an accelerating potential of 20 kV; mineral densities were assumed to be between 8 and 12 g/cm³ (Cabri 1981). Hence, the minimum size of a grain of a *PGM* for fully quantitative analyses was estimated to be 3 μ m to account for the primary excitation area and about 98% of all the fluorescence radiation. In the case of the less dense merenskyite, grains of at least 4 μ m are required. However, all *PGM* observed in the samples were analyzed qualitatively with an energy-dispersion system.

TABLE 1. X-RAY LINES, STANDARDS, LOWER LIMITS OF DETECTION AND REPRODUCIBULITIES FOR MICROPROBE ANALYSES OF Pt-Pd-Bi-Te MINERALS

Line	Standard	LLD	Reproducibility	Range	
Pt <i>L</i> a	PtTe2	n.c.	0.150	up to 38.39	
PdLa	PdB12	n.c.	0.179	0.25-26.10	
TeLα	PtTe2	n.c.	0.309	1.45-57.59	
Bina	PdB12	n.c.	0.516	3.54-79.68	
FeKα	troilite	0.014	0.021	0.12-7.13	
Νίκα	millerite	0.016	0.012	up to 4.14	
CuKa	(Pd,Cu) ₅ Sb ₂	0.018	0.007	up to 1.00	
SbLα	Pds(As,Sb)2	0.034	0.012	up to 1.86	
S Ka	troilite	0.021	0.007	up to 0.44	

LLD: 3-sigma level; reproducibilities: 1 sigma; range: range on which calculations of reproducibilities are based; n.c.: not calculated. Concentrations in weight %.

Quantitative analyses were conducted for Pt, Pd, Te and Bi, as well as for the minor elements Fe. Ni. Cu, Sb and S. In order to obtain the best results for all constituents, it was decided to analyze grains for the major and trace elements in separate sessions. For both sessions, a beam current of 5 \times 10⁻⁸ A was used, measured and monitored on a Faraday cup. Counting times were 20 and 50 seconds at peak position for the major and trace elements, respectively, and 10 seconds for symmetrical background values. Synthetic PGM standards as well as mineral standards were used. Reproducibilities were calculated on 49 duplicate analyses using the formula given by Kaiser & Specker (1956). The lower limits of detection and reproducibilities, as well as the Xray lines and standards used, are given in Table 1.



FIG. 3. Photomicrograph of michenerite (mi), merenskyite (me), and moncheite (mo) with chalcopyrite (cp) and a little pentlandite (ptl) as inclusions in silicate.



FIG. 4. Photomicrograph of moncheite (mo) and michenerite (mi) with chalcopyrite (cp) as inclusions in silicate.

All analyses were performed in duplicate and later averaged. For all calculations, only analyses with totals between 98.0 and 102.0 wt.% were used.

For full ZAF corrections, the program FZAFM supplied by JEOL was used. Furthermore, interelement interferences of Bi with Pt, Te with Sb, and Pt with S had to be considered. Analyses of synthetic standards showed that every 1 wt.% of Bi, Te and Pt resulted in the simulation of 240 ppm Pt, 10 ppm Sb and 5 ppm S, respectively. Use of the BiL α line instead of Bi $M\alpha$ did not improve this situation, as Pt was found to interfere with Bi (1% Pt simulates 900 ppm Bi). Elevated concentrations of S (up to 0.44%) were only encountered in PGM hosted by pyrrhotite and chalcopyrite and, hence, are believed to be caused by interference from the sulfide host. Similarly, Fe and Cu concentrations in these PGM also can be attributed in part to radiation generated in the base metal sulfides. Therefore, compositions of PGM hosted by pyrrhotite or chalcopyrite were corrected for Fe and Cu proportional to the S concentration determined by analysis, with the different sizes of excitation areas of S. Fe and Cu taken into account.

RESULTS

A total of 76 grains of *PGM* were found in eight polished sections from four of the seven samples collected. All grains are very small. The longest axis varies from 1 to 40 μ m, with 62% of the *PGM* being smaller than 4 μ m, and only 9% larger than 10 μ m. Five individual species were observed. Four species, represented by 72 grains, belong to the Pt-Pd-Bi-Te minerals, with the fifth species, sperrylite,

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TABLE 2. REPRESENTATIVE CHEMICAL COMPOSITION AND COMPOSITIONAL RANCE OF MERENSKYITE, MICHENERITE, MONCHEITE AND FROODITE FROM THE UPPER ZONE OF THE EASTERN BUSHWELD COMPLEX

									c.	Total
	Mineral	Pd	Pt	Fe	Ni	Cu	1e	51		
1.	Merenskyite	18.61	10.95	0.95	1.02	0.58	50.42	18.38	n.đ.	100 .9 1
range	(N=3)	18.61-21.23	4.72-10.95	0.95-2.21	0.15-4.14	0.17-0.77	50.42-56.74	13.26-18.38	up to 0.11	
2.	Michenerite	23.03	2.78	0.12	n.d.	0.10	29.85	43.82	0.47	100.17
range	(N=13)	17.65-25.65	up to 10.09	0.12-1.91	up to 0.24	up to 1.00	26.62-31.79	40.13-47.82	0.05-0.81	
3.	Moncheite	9.45	25.99	0.16	0.31	0.19	45.59	18,42	0.17	100.28
range	(N=5)	0.25-9.45	22.95-38.39	0.16-2.47	0.15-1.36	0.19-0.37	37.78-56.20	8.56-21.43	up to 1.10	
4.	Froodite	19.97	n.d.	0.18	n.d.	0.06	1.45	79.68	n.d.	101.34

1. $(Pd_{0.69}Pt_{0.22}Fe_{0.07}Ni_{0.07}Cu_{0.04}) = 1.09(Te_{1.56}Bi_{0.35}) = 1.91$

(Pt_{0.59}Pd_{0.39}Fe_{0.01}Ni_{0.02}Cu_{0.01}∑=1.02</sub>(Te_{1.58}Bi_{0.39}Sb_{0.01}∑=1.98
(Pd_{0.96}Fe_{0.02}∑=0.98^{(Bi}1.96^{Te}0.06∑=2.02

2. (Pd_{0.95}Pt_{0.06}Fe_{0.01}Cu_{0.01}≿=1.03(Bi_{0.92}Sb_{0.02}≿=0.94^{Te}1.03 N = number of analyses (electron-microprobe); n.d. = not detected. Concentrations in weight %.



FIG. 5. Compositions of natural Pt-Pd-Bi-Te minerals from the upper zone (dots) in a (Pd + Pt + Fe + Ni + Cu) - (Bi + Sb) - Te diagram. Shaded areas represent the compositional fields of the respective minerals reported in the literature and illustrate the extent of solid solution between merenskyite and michenerite and in kotulskite.

represented by only four grains. Three grains of electrum also were found. Because of their dominance, the Pt-Pd-Bi-Te minerals were investigated in detail by quantitative electron-microprobe analysis, although grain-size restrictions and the spatial resolution of the electron microprobe allowed quantitative analyses of 33 of these PGM only. Of these, 18 grains were identified as michenerite, 10 grains as moncheite, 4 grains as merenskyite, and one grain as froodite. About 83% of all PGM studied are hosted by silicates, 8% have a common boundary with base metal sulfides, and only 9% occur as inclusions in the sulfides. Two illustrations of the occurrence of Pt-Pd-Bi-Te minerals are shown in Figures 3 and 4. Of the grains in contact with sulfides, eight are associated with chalcopyrite, and five with pyrrhotite. No association of one specific PGM with a specific base metal sulfide could be observed. Commonly, the PGM are accompanied by a number of Bi-Te and



FIG. 6. (Pt+Pd+Fe+Ni+Cu) - (Bi+Sb) - Te diagram showing the compositional variation of moncheite from the upper zone (dots) and the extent of moncheiteinsizwaite solid solution as reported in the literature (hatched area).



FIG. 7. Pd – Pt – (Te+Bi+Sb) diagram showing the compositional variations of merenskyite and moncheite from the upper zone (dots) compared with data from the literature (shaded areas), illustrating complete solid-solution between these two minerals.

Ag-Te minerals. As these minerals are only 1 to 2 μ m in size, they are too small for quantitative analyses by electron microprobe.

Representative compositions and compositional ranges for michenerite, merenskyite, moncheite and froodite in the upper zone are given in Table 2. Figures 5 to 7 show the position of all compositions in the triangular diagrams (Pd + Pt + Fe + Ni + Cu)-(Bi+Sb) - Te and Pd - Pt - (Te+Bi+Sb). Results of analyses with totals between 98.0 and 95.0, as well as between 102.0 and 103.5 wt.%, are included in these diagrams, but are identified by smaller symbols. Also shown in the plots are the compositional variations of merenskyite, michenerite, moncheite, froodite, kotulskite and insizwaite, as given in the literature. These include 41 compositions of merenskyite, 35 of michenerite, 77 of moncheite, 9 of froodite, 64 of kotulskite and 8 of insizwaite taken from Kingston (1966), Mihálik et al. (1974), Cabri & Laflamme (1976, 1981), Watkinson et al. (1978), Cabri et al. (1979), Kingston & El-Dosuky (1982), Volborth et al. (1986), Tarkian (1987), Halkoaho (1989), and Huhtelin (1989). It must be noted that Cabri & Laflamme (1976) reported one composition of merenskyite that plots in the compositional field of michenerite (Fig. 5), which implies an overlap in composition between these two PGM. The separate moncheite field in Figure 6 shows four compositions of moncheite reported by Kingston (1966).

All four Pt-Pd-Bi-Te minerals found in the upper zone contain variable concentrations of Pt, Pd, Bi and Te. In accordance with data from the literature, the upper zone michenerite defines a fairly small compositional field. Merenskyite has an average Pt concentration, but is Bi-rich compared to data from the literature. The composition of moncheite from the upper zone varies most in terms of Pt/Pd and Te/Bi ratios, but all compositions fall within the ranges reported in the literature. It is obvious from Table 3, which lists the Spearman correlation matrix

TABLE	3.	SPEA	RMA	N (CORE	RELATI	ON	MATRIX	FOR	ATOMIC
PROPO	ORTI	ONS	OF	UP	PER	ZONE	Pt-	-Pd-Bi-T	'e Ml	NERALS

-	Pd	Te	Bí	Pt/Pd	Te/Bi
Pt	-0.9398 0.0001	0.5757 0.0003	-0.5845 0.0002	0.9896 0.0001	0.5645 0.0004
Pd		-0.6431 0.0001	0.6756 0.0001	-0.9600 0.0001	-0.6518
Те			-0.9479	0.6317	0.9835
Bí			0.0001	-0.6495	-0.9824
Pt/Pd				0.0001	0.6289

Correlation matrix based on 35 analyses. Error probabilities given underneath the correlation coefficients. for major elements in upper zone Pt-Pd-Bi-Te minerals, that Pt and Te correlate well with Pd and Bi, respectively. In contrast, correlations between the *PGE* and Te or Bi are poor.

All Pt-Pd-Bi-Te minerals were analyzed for As as well. With very few exceptions, all As concentrations were found to be below the lower limit of detection (520 ppm). Traces of As were detected in only one moncheite and two michenerite grains. However, it could not be established whether this is due to a substitution of As for Bi or Te, or to excitation of an As-bearing mineral (*e.g.*, sperrylite) below the exposed surface.

DISCUSSION

The Bushveld Complex is well known for its extensive PGE mineralization at several stratigraphic horizons in the critical zone (e.g., Merensky Reef, UG-2), from which a large variety of different PGM have been described in the literature (e.g., Kingston 1966, Vermaak & Hendriks 1976, Kingston & El-Dosuky 1982). In the upper zone, elevated concentrations of the PGE have so far been documented only from the locality where the samples for this investigation were collected (Von Gruenewaldt 1976, Page et al. 1982). The very low PGE concentrations in many other sulfide-bearing upper zone rocks led Page et al. (1982) and Harney et al. (1990) to conclude that the conditions leading to the formation of the Fe-rich late differentiates of the Bushveld Complex were not favorable for the precipitation of PGE-enriched magmatic sulfides. To date no PGM have been described from the upper zone.

The PGE partition strongly into sulfide melt, with distribution coefficients widely considered to be in the order of thousands (Campbell & Barnes 1984, Naldrett & Barnes 1986). Consequently, any PGM present are expected to be hosted by the base metal sulfides that crystallized from such a sulfide melt. However, almost all PGM occur as inclusions in silicates, without visible contact with base metal sulfides. To explain this textural peculiarity on the assumption of a magmatic process of concentration, selective dissolution of the base metal sulfides that host the PGM is required. The generally high tenor of base metal sulfides in the investigated samples makes it unlikely that such a selective dissolution did occur.

We therefore suggest that these *PGM* formed from hydrothermal fluids, as this could also explain the lateral restriction of the mineralization and the inhomogeneous distribution of the *PGM*, *i.e.*, within a few meters of outcrop, mineralized and unmineralized samples were found. Further support for a hydrothermal origin is provided by alteration and replacement textures of silicates, sulfides and titaniferous magnetite in some samples, as described above. Such an interpretation also would explain the high proportion of chalcopyrite relative to pyrrhotite in mineralized samples. In 100% sulfide of his *PGE*-enriched material, Von Gruenewaldt (1976) found 55% chalcopyrite, 31% pyrrhotite, 6% pentlandite and 8% pyrite. These findings were confirmed in this investigation. In contrast to this modal abundance, the sulfide assemblage in *PGE*-free material from the same locality (Z-2; Harney *et al.* 1990) consists of 63% pyrrhotite, 36% chalcopyrite and 1% pentlandite.

A further indication in favor of hydrothermal activity is the common occurrence of pyrite, as individual inclusions or small veinlets in the silicates, and the presence of sphalerite. The latter is unlikely to form from a sulfide melt, owing to the very low partition-coefficient of Zn into such a liquid (0.5; MacLean & Shimazaki 1976).

Hoffman & MacLean (1976) investigated the ternary system Pd-Te-Bi, with special emphasis on the minerals michenerite and merenskyite. They found that the melting temperature of michenerite is strongly dependent on the Te and Bi concentrations, and varies from 489° (Bi-rich) to 501°C (Te-rich). Furthermore, they observed that in the temperature range 380° - 490°C, michenerite coexists with a) a wide range of BiTess compositions, b) Bi-rich kotulskite, c) Pd- and Bi-rich merenskyite, and d) Te-rich, Pd-deficient froodite. The latter two minerals also have been found to coexist with michenerite in the upper-zone samples. Although upper-zone michenerite contains some Pt (Table 2), the effect of which on the thermal stability of michenerite is not known, the melting temperature and paragenesis of michenerite give a reasonable indication of an upper-temperature limit for the hydrothermal fluids that precipitated michenerite in the footwall of LL2.

This temperature limit could possibly be further constrained with the help of the thermal stabilities imposed by Bi-Te and Ag-Te minerals associated with the *PGM*. Although these minerals are too small for quantitative analyses by microprobe, the data known from the phase diagrams Bi-Te and Ag-Te (Hansen & Anderko 1958) are in agreement with the upper limit in temperature inferred from the stability of michenerite.

A lower limit of the hydrothermal event can be deduced from the composition of plagioclase in the mineralized zone, using the phase diagram for the system albite – anorthite proposed by Smith (1974). Below 400°C, all plagioclase between approximately An_2 and An_{96} should be unstable and ought to break down to albite and a Ca-bearing phase. According to the diagram of Smith (1974), this Cabearing phase should be body-centered anorthite, but in practice epidote \pm calcite forms. The plagioclase from the mineralized samples in the upper zone does

not seem to have undergone this reaction. Its composition (An₆₀ to An₆₃) is indistinguishable from that of plagioclase outside the area of *PGM* mineralization (An₅₈ to An₆₄), which indicates that the temperature of the mineralizing event was in excess of 400°C.

The composition of merenskyite, michenerite and moncheite from the upper zone is highly variable with respect to both the Pt/Pd and Te/Bi ratios (Figs. 5-7, Table 2). A Spearman correlation matrix based on 35 compositions of all Pt-Pd-Bi-Te phases (Table 3) indicates 1:1 substitution between Pt and Pd, and also between Te and Bi. For the individual Pt-Pd-Bi-Te minerals, similar correlations exist. However, the small number of analyses of each species prevents a rigorous evaluation. The observed substitutions confirm the well-documented partial solid-solution series between merenskyite and michenerite, and between moncheite and insizwaite, and the complete solid-solution series between merenskyite and moncheite (Mihálik et al. 1974, Hoffman & MacLean 1976, Tarkian 1987, Grønlie 1988). On the other hand, coupled substitution of the type Pt + Bi for Pd + Te or Pt + Te for Pd + Bi, and vice versa, is not very common, as is indicated by the poor correlations between the PGE and Te or Bi (Table 3).

CONCLUSIONS

Merenskyite, michenerite, moncheite and froodite have been discovered in the sulfide-bearing, anorthositic footwall of LL2 of the upper zone in the eastern Bushveld Complex. Quantitative analyses confirm the solid-solution series known from the literature, and indicate substitution between Pt and Pd, as well as between Te and Bi. Textural and compositional evidence points to precipitation of the *PGM* from hydrothermal solutions at a temperature between 400° and 490°C. Although no stratigraphic horizon continuously enriched in *PGE* was discovered in the upper zone so far, the mineralization described here is evidence that hydrothermal activity in the Fe-rich differentiates of the Bushveld Complex can lead to the local concentration of Pt and Pd.

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